A Review Of Approaches For Setting Acidic Deposition Limits In Alberta

Alberta
ENVIRONMENT

A REVIEW OF APPROACHES FOR SETTING ACIDIC DEPOSITION LIMITS IN ALBERTA

ALBERTA ENVIRONMENT

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FOREWORD

In 1987, a working committee was struck within Alberta Environment to examine the feasibility of implementing a critical loading for acidic deposition based on sulphate and other deposition criteria. The composition of the committee was as follows:

CHAIRMAN	Mr. S.L. Dobko
SECRETARY	Mr. C.S. Liu
MEMBERS	Dr. G. Singleton
	Mr. H. Bertram
	Dr. Y. Lau
	Dr. T. Roberts
	Mr. R. Angle
	Mr. D. Trew
	Ms. B.L. Magill

Dr. S. Goudey, HydroQual Consultants, Calgary, prepared a draft report consolidating the various components of the committee's work. This final report incorporates the editorial comments and consensus of opinions of the committee. Meliza Roberto and Lynn Lockhart typed the document.

EXECUTIVE SUMMARY

The objectives of this report are to summarize previous work on acidic deposition in Alberta and to assess different approaches for setting deposition limits.

Empirical models were used to assess soil and surface water sensitivities to acid loadings (Turchenek and Abboud 1988; Singleton et al. 1988; Erickson 1987; Trew 1988). Sensitive soils were characterized by low base saturation, low cation exchange capacity, and low pH. Surface waters with low alkalinity were considered to be highly susceptible to acidification. The most sensitive areas in Alberta are located on the Canadian Shield (northeastern corner of the province), in certain areas of the Rocky Mountain National Parks, and on organic soils in certain upland regions in the northern part of the province. Generally, sensitive soils and surface waters were found within the same regions. The sensitivities of peatlands and brown water lakes to acidic deposition are not fully understood and require further investigation.

The ionic composition and quantity of acidic deposition in Alberta are unlike those of other regions of Canada, eastern North America, and Europe. Hence, the assumptions and modelling approaches used to assess acid loadings and acidification impacts in these regions are not directly applicable. "Effective Acidity" and "Acidifying Potential", expressed in terms of hydrogen ion equivalents, are two concepts more appropriate than wet sulphate for expressing acid loadings to the environment. With these approaches, more sinks and sources of hydrogen ions can be considered in deriving total mass loadings. Both approaches incorporate elements representing acidic deposition characteristics and receptor response. However, both approaches require more refinement.

Preliminary deposition limits were derived based on the models of Bloom and Grigal (1985) and Dupont and Grimard (1986), which were modified for application to Alberta by Turchenek and Abboud (1987) and Trew (1988), respectively. The range of loadings [$keq(H^+) \cdot ha^{-1} \cdot y^{-1}$] derived for three sensitivity classes of soils and surface waters were:

Sensitivity Class	Soils	Surface Waters
low	0.7 to 1.0	> 0.3
medium	0.3 to 0.4	> 0.3
high	0.1 to 0.3	0.1 to 0.3

Due to the limited range of lake sensitivities incorporated in the Dupont and Grimard (1986) model, deposition limits for surface water could only be derived for the highly sensitive category.

A 10% reduction in base saturation was used to derive loadings for soils. The period of loading was taken as 100 years. A minimum desirable pH of 6.0 was used as the criterion for the determination of deposition limits to surface waters. Empirical observations of damaging acidic deposition in eastern North America support the interpretation that the upper limits of the acid loading ranges proposed for highly sensitive surface waters and soils (0.3 $keq(H^+) \cdot ha^{-1} \cdot y^{-1}$) should not be exceeded. Naturally acidic aquatic systems (i.e., brown waters) were not considered in the establishment of loadings. A more detailed spatial assessment of ecosystem sensitivity and establishment of regional deposition limits for acidic deposition should be considered.

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1. INTRODUCTION

"Acid Rain" is an important environmental issue of public concern in Alberta. Acid rain is defined here as the deposition of acid and acid precursors in precipitation (wet deposition: rain, snow, fog) and as gases, aerosols, and solids (dry deposition). Long-term inputs of acidic and acid-forming substances can alter the structure and function of aquatic and terrestrial ecosystems. The ecological and socio-economical consequences of acidic deposition in Europe and regions of eastern North America are well documented (references in Legge and Crowther 1987; Legge 1988a,b).

The prediction of acidic deposition impacts on aquatic and terrestrial ecosystems is difficult because the effects are exceedingly complex, subtle, and long-term. Regulators are faced with an equally difficult task of establishing acceptable emission and deposition limits to preserve environmental integrity.

All ecosystems have an inherent capacity to resist acidification. Thus, a realistic management objective is to balance acidic loadings with the sensitivity of a particular ecosystem to acidic deposition. The establishment of deposition limits based on ecosystem sensitivity is part of the management strategy developed for western and northern Canada (Sandhu and Wilson 1987).

In 1987, the Western and Northern Canada Long Range Transport of Atmospheric Pollutants Technical Committee commissioned the Western and Northern Canada Interim Acid Deposition Target Loadings Task Group to establish acceptable interim target (critical) loadings of acidic deposition for the four western provinces, the Yukon, and the Northwest Territories. Each jurisdiction was asked to review the pertinent aspects

of the acidification issue and derive appropriate deposition limits. In Alberta, preliminary evaluations of precipitation chemistry and present loadings to lakes and soils have been completed (Lau 1982; Palmer and Trew 1987; Holowaychuk and Fessenden 1987). Empirical models have also been applied to assess acidification impacts and scope out deposition limits for soils (Turchenek and Abboud 1988; Singleton et al. 1988) and surface waters (Erickson 1987; Trew 1988).

The objectives of this report are to summarize previous work on acidic deposition in Alberta and to discuss two new approaches for setting limits and measuring acidic deposition. "Effective acidity" (EA) is quantified as the sum of hydrogen ion equivalents present in precipitation and those which could be generated through biological and chemical transformations of "acid-forming substances" in wet and dry deposition, minus all potential losses from processes that consume hydrogen ions (base reactions). "Acidifying potential" is an alternate concept for expressing acidic deposition which corrects for non-acidic sulphates and sea-salt in precipitation and includes a factor for nitrate leaching from watersheds (Brydges and Summers 1989). The rationale and technical justification for applying these approaches to Alberta, as opposed to approaches based on "wet sulphate" modelling, are discussed in this document.

This report is not intended to be a comprehensive survey of the literature. The published material on acidic deposition is vast and has been the subject of numerous reviews (see references in Legge 1988b). Nor is it intended to represent Alberta's final position on acidic deposition limits. It is anticipated that the data and approaches presented herein will be modified and refined in the future.

A discussion of the general nature of acidification impacts on terrestrial and aquatic ecosystems is presented first. Previous work quantifying the sensitivities of Alberta lakes and soils is then summarized, followed by a discussion of precipitation chemistry in Alberta. Methods to model loadings and prediction of acidification impacts are discussed briefly, and deposition limits are then proposed for Alberta. Information needs for management and additional concerns are addressed in the final section.

Terms pertinent to the discussion of acidic deposition and the derivation of deposition limits are defined in the Glossary of Terms in the Appendix.

2. GENERAL ACIDIC IMPACTS ON AQUATIC AND TERRESTRIAL ECOSYSTEMS

The impacts of acidic deposition on soils and aquatic ecosystems were reviewed by Telang (1987), Turchenek et al. (1987a), and Legge and Crowther (1987). Symptoms of ecosystem damage from acidification are summarized in Tables 1 and 2. The gradual accumulation of hydrogen ions within aquatic and terrestrial ecosystems produces subtle changes in ecosystem structure and function. The rate of acidification is a complex function of ecosystem sensitivity (resilience) and the quantity, acidity, and ionic composition of deposition.

Acidified aquatic ecosystems are characterized by decreased species diversity and a more simplified community structure (reviewed in Telang 1987). Fish populations are usually depleted as a result of reproductive failure, altered growth, or direct mortality. The diversities of zooplankton and phytoplankton populations are reduced with

Table 1. Potential effects of increasing acidity on aquatic ecosystems.

PARAMETER		RESPONSE Increase; Decrease
species diversity	-bacteria	D
	-macrophytes and algae	D
	-invertebrates	D
	-fish (plus other vertebrates)	D
species abundance	-bacteria	D
	-macrophytes and algae	D
	-invertebrates	D
	-fish (plus other vertebrates)	D
productivity (all trophic levels)		D
microbial activity		D
sediment nutrient recycling		D
food chain complexity		D
metal bioavailability and potential toxicity		I
alkalinity, acid neutralizing capacity		D
mineral acidity		I

D = Decrease
I = Increase

General References: Erickson (1987)
Seip et al. (1986)
Telang (1987)

Table 2. Potential impacts of acidic deposition on soil systems.

PARAMETER	RESPONSE Increase; Decrease
cation exchange capacity	D
exchangeable acidity	I
base saturation	D
weathering process (dissolution)	I
organic matter decay	D
organic matter production	D
soil integrity (organo-mineral associations)	D
nutrient availability	D
leaching of most plant nutrients	I
metal solubility, availability, potential toxicity (particularly aluminum)	I
microbial, plant, invertebrate diversity (shift to acid/metal tolerant species)	D
microbial ammonification/nitrification	D
release of reduced sulfur and carbon to the atmosphere	I

D = Decrease
I = Increase
General References: Holowaychuk and Fessenden (1987)
Reuss and Johnson (1986)
Turchenek et al. (1987a)
Seip et al. (1986)

a shift to more acid-tolerant species. Similar trends are observed for other life forms including benthic invertebrates, attached aquatic plants, and bacteria.

Elevated hydrogen ion concentrations may directly affect individual species. Alternatively, changes in community structure as a result of acidification may indirectly affect other species. Changes in microbial populations, for example, can alter nutrient recycling dynamics and thus interfere with phytoplankton growth and possibly other trophic levels. The increased solubilities of potentially toxic trace metals (i.e., aluminum) at low pH conceivably could have similar direct and indirect effects. Acidic deposition impacts on aquatic ecosystems are complex, and cause and effect relationships are difficult to define.

The sensitivity of aquatic ecosystems to acidification is determined largely by physical, chemical, and biological characteristics of the watershed (Henriksen 1982) and waterbody (Telang 1987; Schindler 1986). Acidic substances are deposited directly into the waterbody or reach the system through the watershed. Reactions within the soil and with vegetation can alter the ionic composition and quantity of acidic loadings (Reuss 1975). Land use patterns (forested versus agricultural lands) will also alter the nature of interaction between the watershed and chemical constituents of precipitation.

Sensitive waterbodies are often shallow, low volume systems with a low watershed to lake surface area ratio. Water quality conditions in such systems may be determined, in a more direct sense, by atmospheric deposition with little moderating influence from the drainage basin.

The principal buffer system in most freshwaters is based on the carbonic acid-bicarbonate equilibrium. Organic substances and

metal hydroxides can be effective (and dominant) buffers at lower pH ranges in naturally acidified, brown water systems. Alkalinity, the capacity to neutralize hydrogen ions, can be defined as:

$$[Alk] = [HCO_3^-] + [CO_3^{2-}] + [CO_3^{2-}] + [OH^-] + [A^-] - [H^+]$$

where A^- represents organic anions. In solutions where organic and metal components contribute to buffering, the buffering quantity is referred to as the acid neutralizing capacity (ANC).

Precipitation entering the soil undergoes numerous reactions. Plant nutrients such as nitrate, ammonium, and sulphate are transformed and/or cycled through the biota generating or removing hydrogen ions. Hydrogen ions also exchange with metal cations on soil binding sites. These cation exchange reactions buffer the soil solution against changes in pH. However, the pH will drop drastically once the cation exchange capacity of a soil is exceeded. Leaching of cations is accompanied by a loss of anions to maintain electroneutrality. Losses of mobile anions such as nitrate and sulphate increase with increasing acid loadings.

Colloidal organic and inorganic materials can also contribute to the buffering capacity of soils. The hydrolysis of aluminum species, for example, is an important buffering mechanism in certain acidic soils (pH 3.5 to 5.5). Humic substances and other organic matter contain numerous functional groups that can participate in proton transfer reactions, hence buffer against pH changes.

The sensitivity of soils to acidification is determined by the relative contribution of the different mechanisms regulating the soil

solution hydrogen ion concentration. Soils with a low cation exchange capacity or buffering capacity would be considered susceptible to acidification.

The nature of acidification impacts on terrestrial ecosystems is similar to aquatic ecosystems. A decrease in species diversity occurs with a shift to more acid-tolerant species. Accelerated leaching of nutrients limits above ground production. Solubilization of toxic metals such as aluminum, in addition to hydrogen ion, may adversely affect the biota. Other characteristics of acidified soil systems are summarized in Table 2.

2.1 SENSITIVITY OF ALBERTA LAKES AND SOILS TO ACIDIC DEPOSITION

The sensitivities of soils and surface waters to acidic deposition in the four western provinces and the Northwest Territories were assessed through systematic regional surveys coordinated by the Western and Northern Canada Long Range Transport of Atmospheric Pollutants Technical Committee. Results from the sensitivity mapping of soils (Holowaychuk and Fessenden 1987) and surface waters (Erickson 1987) in Alberta are summarized by Palmer and Trew (1987). Sensitivity is a measure of the potential susceptibility of the ecosystem to acidification as determined by selected inherent physical and chemical characteristics.

The parameters used to assess the sensitivity of aquatic systems to acidic deposition (National Research Council of Canada 1981) included alkalinity, calcium, and pH (Table 3; Erickson 1987) although alkalinity is the most useful index. Surface waters in southern and central Alberta

Table 3. Criteria for evaluating the sensitivity of Alberta lakes to acidification (Erickson 1987).

Sensitivity Rating	Parameter Range		
	Alkalinity (mg/L)	Calcium (mg/L)	pH
High	0 to 10	0 to 4	<6.5
Moderate	11 to 20	5 to 8	6.6 to 7.0
Moderate-Low	-	9 to 25	-
Low	21 to 40	26 to 40	7.1 to 7.5
Least	>40	>40	>7.5

were found to be well buffered and alkaline (largely due to the carbonate-rich surficial deposits and bedrock geology). Hence, these regions are relatively insensitive to acidic deposition. Lakes in specific regions of the Rocky Mountain National Parks, the Canadian Shield area, and certain northern upland regions were assessed as highly sensitive to acidic deposition. Highly-coloured, muskeg-type lakes in the Caribou Mountains Uplands and Birch Mountains Uplands were also classified as sensitive because they have little bicarbonate alkalinity. However, many of these systems are naturally acidic and may be buffered in a lower pH range by organic acids or metal hydroxides, as discussed previously. These characteristics of brown water lakes require further evaluation in order to assess their actual sensitivities to acidification, and to derive appropriate deposition limits.

Two assessments of terrestrial sensitivity were undertaken. First, the potential of soils and the underlying bedrock to reduce the acidity of atmospheric deposition before entering into aquatic ecosystems was evaluated, and, second, the sensitivity of topsoil to acidic inputs and the effects on its ability to sustain plant growth were assessed (Holowaychuk and Fessenden 1987).

Soil depth, exchangeable bases, the type of bedrock and parent material, drainage class, and type of surficial material were criteria used to rate the potential of soils to reduce acidity (Table 4). Deep soils with a high exchangeable base content have a high potential to reduce acidic inputs irrespective of the underlying bedrock. The type of bedrock has a greater influence on shallower soils. Roughly 65% of Alberta is covered by soil and geological materials with a high potential

Table 4. Criteria for potential of Alberta soils and geology to neutralize acidic inputs (Holowaychuk and Fessenden 1987).

POTENTIAL TO NEUTRALIZE	SOIL DEPTH (cm)	EXCHANGEABLE BASES (cmol(+)/kg)	BEDROCK TYPE[1]	SURFICIAL MATERIAL TYPE[2]
high	all depths	calcareous	all types	all types
	<25	all ranges	I	A
	25 - 100	>15	I, II, V	A
	25 - 100	6 - 15	I, II	A
	25 - 100	<6	I	A
	>100	>15	all types	all types
	>100	6 - 16	I	A
	Poorly drained soils	all ranges	all types	all types
	Eutrophic peatlands	-	-	-
medium	<25	all ranges	II, III	-
	25 - 100	6 - 15	II, III	B, C
	25 - 100	<6	II, III	B
	>100	6 - 15	II, III, IV	B, C, D
	>100	<6	I	A
	Mesotrophic peatlands	-	-	-
low	<25	all ranges	IV	-
	25 - 100	<6	IV	C, D
	>100	<6	II, III, IV	C, D
	Oligotrophic peatlands	-	-	-

[1] Bedrock Types:
- I carbonate or calcareous clastic sedimentary rocks
- II intermingled carbonate and clastic sedimentary rocks
- III clastic sedimentary rocks
- IV granite type rocks
- V alkaline clastic sedimentary rocks

[2] Surficial Material Types:
- A carbonate materials
- B non-carbonate, weak calcareous, clay texture
- C non-carbonate, weak calcareous, fine loamy texture
- D non-carbonate, coarse texture

to reduce acidity. Only 11% is considered to have a low potential (medium potential = 22%). Soils with a low potential to reduce acidity are located within the Rocky Mountain National Parks, Caribou Mountains, Cameron Hills (Grande Prairie region), Canadian Shield (northeast corner of the province), and Birch Mountains. Many of these areas with low potential are associated with mesotrophic peatland systems or with regions of non-carbonate bedrock and sandy soils, with low exchangeable base contents.

The sensitivities of topsoil and peatlands to acidic inputs were assessed based on the following criteria (Table 5; Holowaychuk and Fessenden 1987):

1. Cation exchange capacity;
2. pH;
3. Sensitivity to base loss;
4. Sensitivity to acidification; and
5. Sensitivity to solubilization of aluminum.

Only undisturbed topsoils (top 20 cm) were included in the sensitivity mapping. Agricultural soils and the properties and potential influences of subsoils were not considered. The surface soils play a major role in plant growth. Hence, the sensitivity rating provides some indication of the susceptibility of the soil to chemical changes that could have a deleterious effect on plant growth.

Peatlands were first classified into one of three categories (eutrophic, mesotrophic, oligotrophic) based on available chemical data (soil pH, exchangeable bases, base saturation, pool water pH, and total calcium plus magnesium). The peatland systems separated by category were

Table 5. Criteria for the sensitivity rating of mineral soils and peatlands (Holowaychuk and Fessenden 1987).

	SENSITIVITY TO:											
	Base Loss			Acidification			Aluminum Solubilization			Rating		
Mineral Soils	CEC			CEC			CEC			CEC		
	1	2	3	1	2	3	1	2	3	1	2	3
pH <4.6	H	H	H	L	L	L	H	H	H	H	H	H
4.6 - 5.0	H	M	M	L	L	L	H	H	H	H	M	M
5.1 - 5.5	H	M	M	M	LM	L	H	M	M	H	M	M
5.6 - 6.0	H	M	L	H	LM	LM	M	LM	LM	H	M	L
6.1 - 6.5	H	L	L	H	L	L	L	L	L	H	L	L
>6.5	L			L			L			L		
Peatlands												
eutrophic	L			L			L			L		
mesotrophic	H			H			M			H		
oligotrophic	L			L			H			L		

CEC - Cation Exchange Capacity (cmol (+)/kg)
ranges: (1) <6; (2) 6-15; (3) >15

H - High sensitivity
M - Medium sensitivity
L - Low sensitivity

then evaluated for sensitivity to base loss, acidification, and aluminum solubilization. Eutrophic peatlands were characterized by high pH, high cation exchange capacity, high buffering capacity, and low potential for aluminum solubilization. These systems were considered relatively insensitive to acidic inputs.

Oligotrophic peatland systems are acidic soils with low base saturation, low cation exchange capacity, and low soluble levels of base cations. These systems are buffered by organic acids and metal hydroxides. The potential for aluminum solubilization is high but organic substances may bind and detoxify available metals. Since these systems are naturally acidified, their sensitivity to acidic deposition was considered to be low. Mesotrophic and more transitional systems were considered to be highly sensitive to acid loadings.

On an areal basis, roughly 44% of the province contains soils considered insensitive to acidic inputs, 31% contains soils of medium sensitivity, and 23% contains highly sensitive soils (surface waters account for the remaining area). The most sensitive soils are in areas of non-carbonate rocks (Rocky Mountain National Parks), granite-type rocks (Canadian Shield), peatlands and sandy soils with a low base cation content. The productive agricultural soils in the southern and central regions of the province are relatively insensitive.

The geographic distribution of aquatic ecosystems sensitive to acidic deposition is generally consistent with soil sensitivity, and the potential of soils and geology to reduce acidity. However, unlike other regions of Canada, the relative sensitivities of soils and aquatic systems in Alberta are spatially heterogeneous. This factor may have

implications for the future in terms of establishing regional deposition limits.

3. ACIDIC DEPOSITION

Acidic substances and acid precursors are removed from the atmosphere by wet and dry deposition processes (Figure 1). Wet deposition is the transfer of airborne substances to the Earth's surface in rain, snow, or fog. Dry deposition refers to processes (including impaction, Brownian motion, sedimentation, adsorption, absorption, and chemical reaction) that facilitate the transfer of substances from the atmosphere to the Earth's surface in the absence of water. Oxides of sulphur and nitrogen, and ammonia (NH_3) are three important gaseous components of dry deposition, as is "dust" from wind erosion. However, the impact of airborne alkaline soil on precipitation chemistry in Alberta is not well documented (Schewchuk et al. 1989).

Acid-forming substances in atmospheric deposition produce hydrogen ions when they undergo chemical and biological transformations. Processes that can produce and consume hydrogen ions include weathering, ion exchange, redox reactions, and biologically mediated synthesis and decomposition (Table 6).

3.1 PRECIPITATION CHEMISTRY: BASIC CONCEPTS

Pure water in equilibrium with atmospheric carbon dioxide has a pH of 5.6. The presence of other soluble compounds will increase or decrease this pH based on their aqueous acid-base chemistry. However, the carbonic acid-bicarbonate equilibrium is an effective buffer against

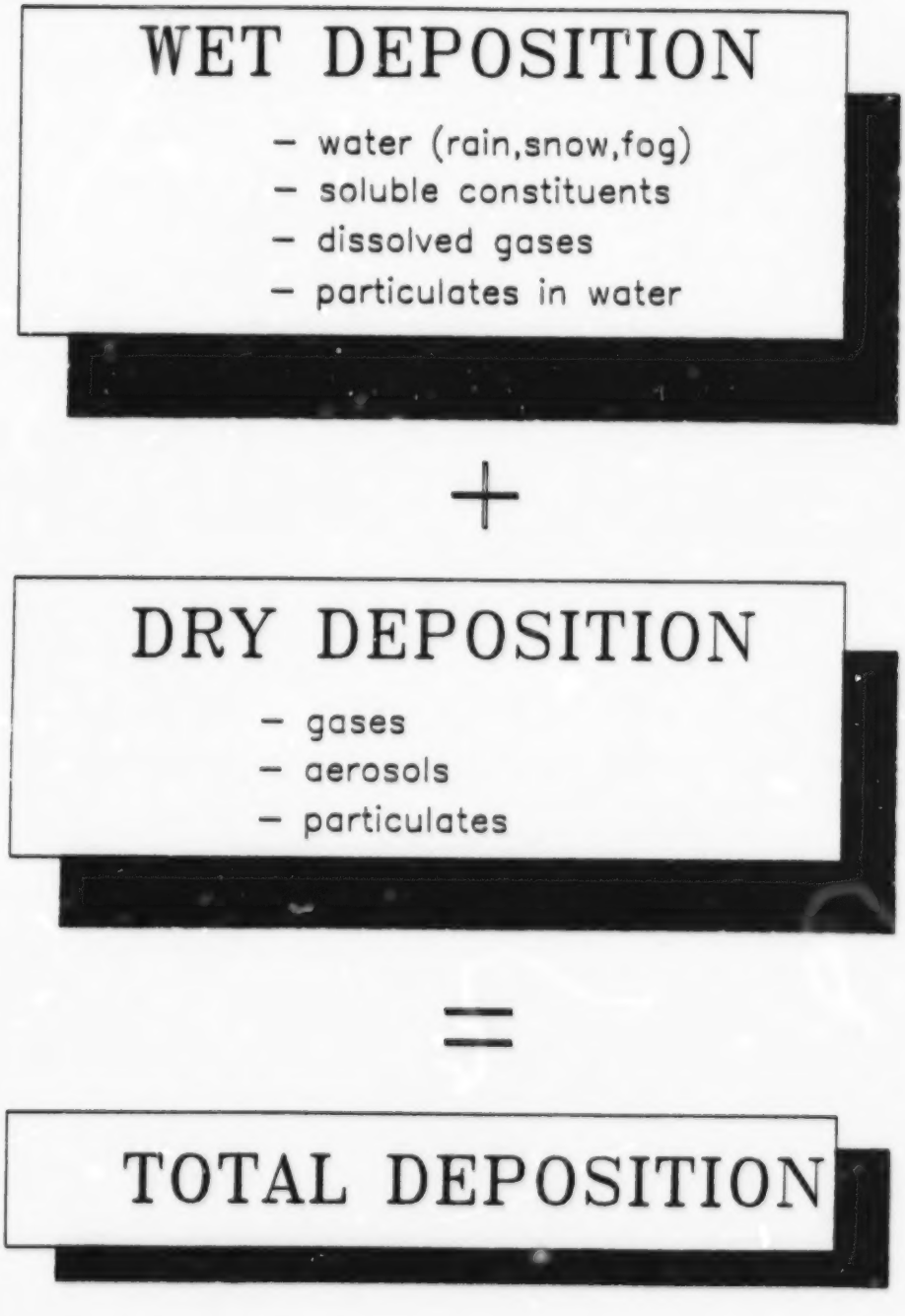

WET DEPOSITION
– water (rain,snow,fog)
– soluble constituents
– dissolved gases
– particulates in water
+
DRY DEPOSITION
– gases
– aerosols
– particulates
=
TOTAL DEPOSITION

Figure 1

Table 6. Processes that can modify the hydrogen ion balance in aquatic and terrestrial ecosystems. (after Gorham et al. 1984)

PROCESS	EQUATION
Weathering	
calcite	$CaCO_3(s) + H^+ = Ca^{2+} + HCO_3^-$
anorhite	$CaAl_2Si_2O_8(s) + 2H^+ = K^+ + 2H_4SiO_2O_5(OH)(s)$
potassium feldspar	$KAlSi_3O_8(s) + H^+ + 4\ 1/2H_2O = K^+ + 2H_4SiO_4 + 1/2Al_2Si_2O_5(OH)_4(s)$
gibbsite	$Al_2O_3 \cdot 3H_2O + 6H^+ = 2Al^{3+} + 6H_2O$
olivine	$MgFeSiO_4(s) + 4H^+ = Mg^{2+} + Fe^{2+} + H_4SiO_4$
albite	$2NaAlSi_3O_8(s) + 2H^+ + 9H_2O = 2\ Na^+ + 4H_4SiO_4 + Al_2Si_2O_5(OH)_4(s)$
Ion-exchange	
Cation exchange	$NaR + H^+ = HR + Na^+;\ CaR_2 + 2H^+ = 2HR + Ca^{2+}$
Anion exchange	$2ROH + SO_4^{2-} = R_2SO_4 + 2OH^-$
Redox (Microbial)	
Nitrification	$NH_4^+ + 2O_2 = NO_3^- + H_2O + 2H^+$
Denitrification	$5CH_2O + 4NO_3^- + 4H^+ = 5CO_2 + 2N_2 + 7H_2O$
Oxidation of H_2S	$H_2S + 2O_2 = SO_4^{2-} + 2H^+$
SO_4^{2-} reduction	$SO_4^{2-} + 2CH_2O + 2H^+ = 2CO_2 + H_2S + H_2O$
Pyrite oxidation	$FeS_2(S) + 3\ 3/4O_2 + 3\ 1/2H_2O = FE(OH)_3 + 2SO_4^{2-} + 4H^+$
Synthesis and Decomposition of Biomass and Humus.	
Photosynthesis with NO_3^- assimilation, anaerobic respiration, NH_4^+ assimilation, anaerobic mineralization (half-reaction) N_2 fixation (half-reaction).	

the development of extreme alkaline conditions in rainfall. Hence, precipitation is normally acidic and basic rain is relatively uncommon.

Precipitation contains a complex mixture of dissolved gases, and inorganic and organic substances. Major sources of atmospheric contaminants in precipitation include volcanic eruptions, biogenic gas production, seaspray, windblown soil dust, and anthropogenic emissions (Legge 1988a; Schewchuk et al. 1989). Strong acids, such as sulphuric and nitric are derived from atmospheric pollutants which originate from the combustion of fossil fuels. Sulphuric acid is formed from oxidation of the combustion product sulphur dioxide (SO_2) and reaction with water:

$$SO_2 + 1/2\ O_2 + H_2O = H_2SO_4 \qquad (1)$$

In rainwater, the sulphuric acid is completely dissociated ($2H^+ + SO_4^{2-}$).

Nitric acid results from the oxidation of nitric oxide to nitrogen dioxide and reaction with water:

$$NO + 1/2\ O_2 = NO_2;\ 3NO_2 + H_2O = 2HNO_3 + NO \qquad (2)$$

Dissociation of the parent acid gives one hydrogen ion and the conjugate base nitrate.

Precipitation acidity is ultimately determined by the reaction of acid generated from atmospheric pollutants w h basic substances such as calcium and magnesium carbonates (alkaline dust). The presence of these basic substances therefore have an important role in

precipitation chemistry (Summers 1986; Schewchuk et al. 1989).

Alkaline dust is derived primarily from soils entrained into the atmosphere by wind action. Hence, regional patterns of land use will determine the relative contribution of this material to precipitation chemistry. Ammonia also participates in acid-base reactions and reduces the acidity of precipitation (ApSimon et al. 1987; Bertram et al. 1987). Major sources of ammonia include feed lots, biological decomposition, and agricultural lands that receive inputs of ammonia and related nitrogen fertilizers (i.e., urea). As with alkaline dust, the relative importance of ammonia in precipitation quality is largely determined by land use patterns. However, although ammonia (NH_3) will ionize to ammonium (NH_4^+) in precipitation and thereby reduce the available hydrogen ion concentration, ammonium may enhance receptor acidification through nitrification (see Table 6 and Section 3.3). The exact proton balance associated with ammonia and nitrate cycling remains the subject of debate.

The chemistry of precipitation is dynamic and it is unlikely that an equilibrium is achieved at the time of deposition on the receptor. Reactions involving dissolved gases, dissolution of solids, or among constituents in precipitation proceed at different rates.

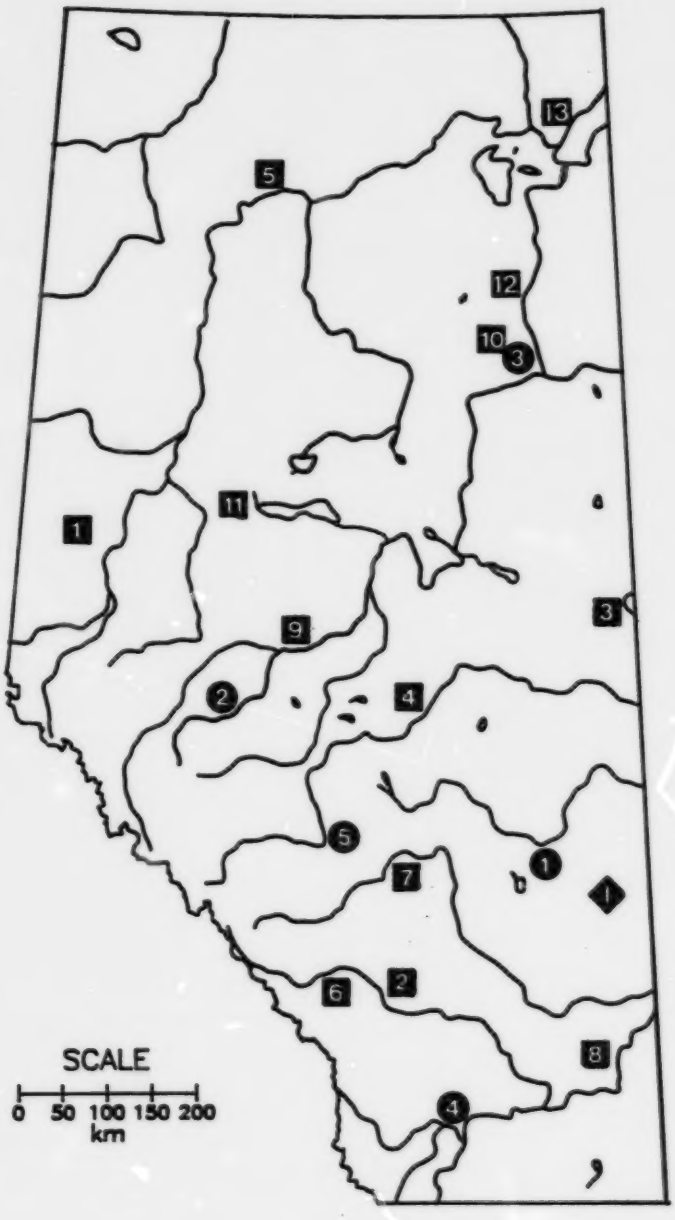
In summary, acids and acid precursors are primarily derived from anthropogenic emissions and basic substances are derived from natural sources. Acid-base reactions determine the ionic composition and acidity of precipitation, which will vary based on land use patterns and proximity to emission sources. Other components in wet deposition are of less importance (sodium and chloride of marine origin) or not well understood (organic substances).

3.2 MONITORING ACIDIC DEPOSITION AND ITS EFFECTS IN ALBERTA

Many of the precipitation monitoring stations in Alberta are near urban centres or point source emissions (Figure 2). Five stations were installed by the Atmospheric Environment Service of Environment Canada in 1978 as part of the Canadian Network for Sampling Precipitation (CANSAP). These stations now have been phased out along with the Air and Precipitation Network (APN) and replaced by the Canadian Air and Precipitation Monitoring Network (CAPMON). A CAPMON station has been established at Esther in southern Alberta. Alberta Environment installed six provincial monitoring sites in 1978 and has installed an additional five stations since 1985: Fort Vermilion, Kananaskis, Cold Lake, Fort McMurray, High Prairie. Stations at Fort MacKay and Fort Chipewyan were added in 1989.

Numerous air quality monitoring stations are also operated by industry as part of their licensing requirements. More recently, the Government/Industry Acid Deposition Research Program (ADRP) completed a two-year (1985 to 1987) intensive sampling program in southern Alberta (Legge 1988b). The objective was to characterize air quality conditions and identify acidification impacts.

The provincial and federal monitoring stations in Alberta only measure wet deposition. Descriptions of the sampling techniques, sites, and equipment are available (Stanley Associates 1978; Barrie and Sirois 1982). The constituents and properties of wet deposition that are routinely measured include pH, conductivity, major cations (calcium, magnesium, ammonium, sodium, potassium), and major anions (sulphate, nitrate, chloride, and phosphate). Bicarbonate concentrations are



● CANSAP
(phased out in 1985)

1 Coronation
2 Edson
3 Ft. McMurray
4 Lethbridge
5 Rocky Mtn House

■ Alberta Environment

1 Beaverlodge
2 Calgary
3 Cold Lake
4 Edmonton
5 High Level (Ft. Vermilion)
6 Kananaskis
7 Red Deer
8 Suffield
9 Whitecourt
10 Ft. McMurray
11 High Prairie
12 Fort Mackay
13 Fort Chipewyan

◆ CAPMoN

1 Esther

daily sampling

Monitoring Stations for Wet Deposition

Figure 2

calculated from the pH of the sample and gas phase equilibria. Charge and conductivity balances are used to assess the completeness of the analysis. If unmeasured ionic species are present, a charge imbalance will occur. Detailed analytical procedures for each parameter are given in the Methods Manual for Chemical Analysis of Atmospheric Pollutants prepared by the Alberta Environmental Centre (1985). Only the dissolved fraction of the total constituent present is actually measured. Hence, the results may under- or over-estimate potential loadings of acidic substances and precursors.

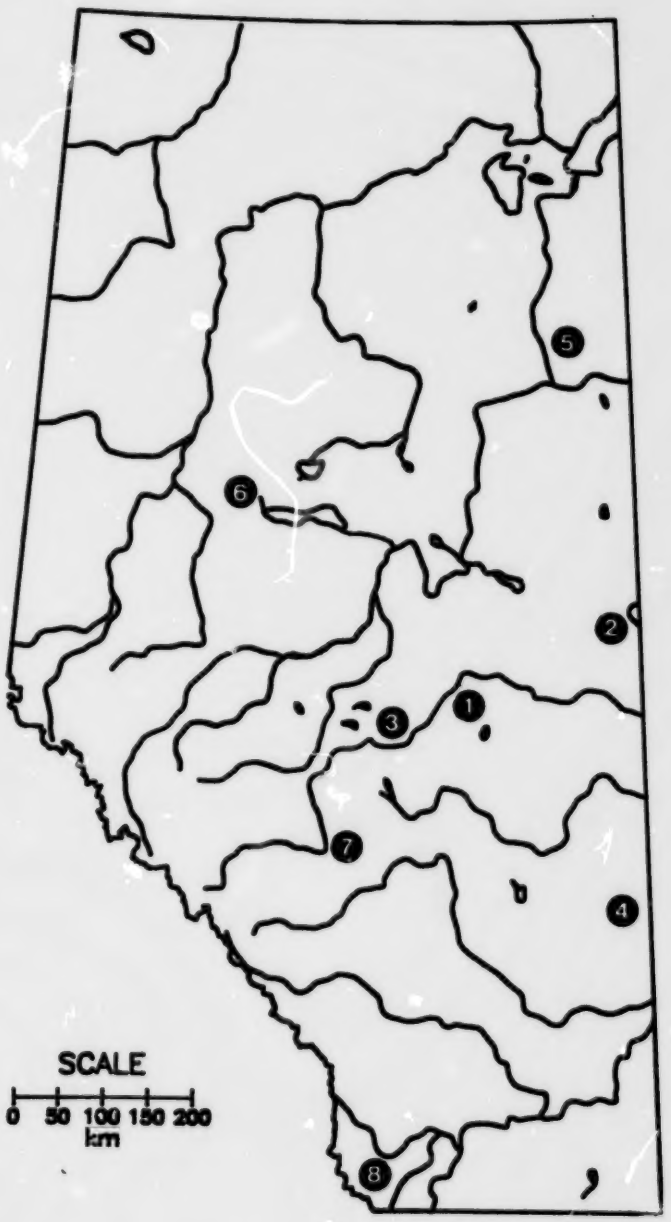
Wet deposition data in Alberta have been analyzed by Barrie and Sirois (1982) and more recently by Lau (1982), Lau and Das (1985), Bertram et al. (1986), Sandhu and Blower (1986), and Legge and Crowther (1987). Precipitation chemistry is highly variable over time and on a regional basis (Lau 1982). During 1978 to 1982, the average pH of precipitation in Alberta was 5.5. The precipitation in southern Alberta generally contains higher levels of calcium and magnesium and a higher pH than precipitation in other regions of the province (Bertram et al. 1986; Lau 1982). During periods when major ion concentrations are low, the hydrogen ion concentration tends to be higher (lower pH). Major changes in precipitation chemistry were not detected over this five-year period. Precipitation volume is also highly variable on a regional and event basis. However, total annual precipitation for Alberta varies in a narrow range from 38 to 58 cm (1978 to 1987).

There is very little available information on dry deposition in Alberta. Alberta Environment operates high-volume samplers for the National Air Pollution Surveillance Network (NAPS) to monitor suspended

particulate matter. Most sites, however, are near urban centres and the collected particulate matter is not routinely analyzed for sulphate and other ions of interest in precipitation chemistry.

In Alberta, the contribution of dry deposition has only been estimated from ambient gas concentrations and calculated deposition velocities (Kociuba 1984, Sirois and Barrie 1986, Legge 1988b). Other common methods for estimating dry deposition, in addition to flux parameterization, include mass balance analyses, and flux monitoring (reviewed by Hicks et al. 1980, 1987). Techniques for measuring dry deposition of sulphur dioxide in Alberta were evaluated by Hunt et al. (1981, 1982) and Chadder et al. (1984). Atmospheric flux measurements (eddy accumulation and concentration gradient profile techniques) were considered the most suitable, particularly for application in an experimental program. More recently, Davis and Hunt (1987) evaluated methods for measuring dry deposition and made recommendations for establishing a regional scale dry deposition monitoring network in Alberta. However, dry deposition is not routinely monitored in Alberta at the present time.

The Soil Protection Branch of Alberta Environment has established eight long-term soil acidification monitoring sites throughout Alberta since 1981 (Figure 3). The program objective is the early detection of acidification impacts on soil fertility and properties that can affect plant growth (H. Regier, personal communication). Parameters that are measured include pH, calcium, magnesium, potassium,



Long Term Soil Acidification Monitoring Sites

Figure 3

sodium, aluminum, iron, manganese, sulphur, nitrogen, carbon, and phosphorus. The sites were selected to permit early detection of subtle changes and for long-term monitoring purposes. The following criteria were considered in selecting appropriate sites:

1. Areas receiving maximum loadings of acidic deposition;
2. High soil sensitivity;
3. Uniform soil characteristics throughout site area;
4. Undisturbed native soils;
5. Site establishment legally secured to assure long-term access.

The Environmental Quality Monitoring Branch (EQMB) of Alberta Environment has initiated a long-term lake monitoring project at three sensitive sites in northeastern Alberta. The purpose of this work is to identify significant changes over time in either the trophic state or acidification status of these waterbodies. Two poorly buffered, unnamed lakes on the Muskeg Mountain Upland have been sampled monthly since May 1988. Legend Lake, on the Birch Mountains Upland, was added during 1989.

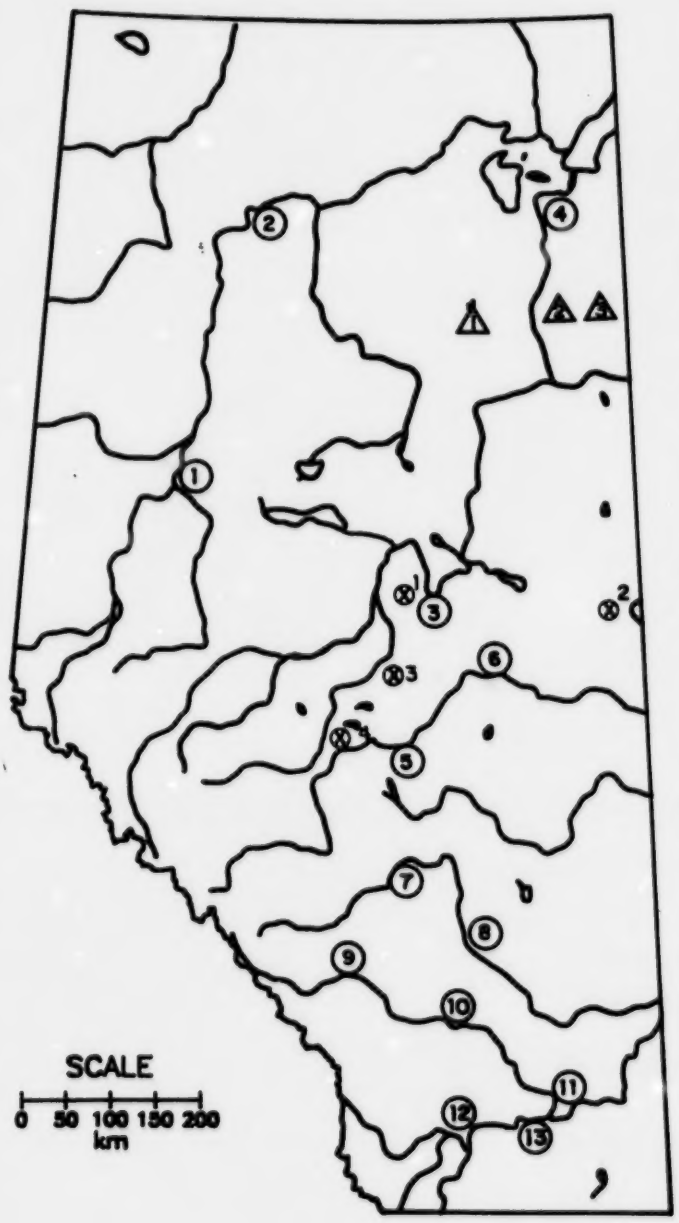
Extensive surveys have also been conducted in this region in recent years in order to validate and upgrade the historical data base which was employed for the original sensitivity mapping. Since 1987, a total of 67 lakes have been sampled during summer and winter. In addition, a preliminary survey to determine the significance of snowmelt pH depression was initiated in 1989 on three streams draining the Muskeg Mountain Upland: Firebag, Muskeg, and Steepbank rivers.

In addition to these priority sensitive zones, the EQMB maintains extensive long-term river and lake monitoring networks throughout Alberta (Figure 4). The sites have been established to monitor the water quality trends resulting from a variety of watershed activities but can also provide a substantial reference data base for any long-term acidification pattern in less sensitive zones.

3.3 THE EFFECTIVE ACIDITY AND ACIDIFYING POTENTIAL CONCEPTS

Wet deposition in the western provinces was compared with other regions in Canada (in particular the lower Great Lakes region [Figure 5]) and eastern North America (see Munger and Eisenreich 1983; Summers 1986; Sandhu and Blower 1986; ESSA and Concord Scientific Corp. 1987; Legge 1988b). It is important to note that volume-weighted mean concentrations of the strong acid anions, sulphate and nitrate, are substantially lower in the precipitation of the western provinces. In addition, the levels of hydrogen ion are also significantly lower and close to the theoretical pH value for pure water in equilibrium with atmospheric carbon dioxide (pH 5.6). The concentration of sodium, chloride, potassium, and ammonium are, however, similar in magnitude between both regions (Figure 5). It is clear from Figure 5 that the relative abundances of strong mineral acids (sulphuric, nitric) are lower in precipitation of the western provinces.

Sulphate and hydrogen are the dominant ions in precipitation of the lower Great Lakes region. Hence, one observes good correlations between wet sulphate and hydrogen ion concentrations, and sulphate is often used as a surrogate for acidity in empirical models. However, this



LONG-TERM RIVER NETWORK

1. SMOKY RIVER AT WATINO
2. PEACE RIVER AT FORT VERMILION
3. ATHABASCA RIVER AT ATHABASCA
4. ATHABASCA RIVER AT OLD FORT
5. NSR AT DEVON
6. NSR AT PAKAN
7. RED DEER RIVER AT RED DEER
8. RED DEER RIVER AT MORRIN
9. BOW RIVER AT COCHRANE
10. BOW RIVER AT CARSELAND
11. BOW RIVER AT RONALENE BRIDGE
12. OLDMAN RIVER AT LETHBRIDGE
13. OLDMAN RIVER AT TABER

LONG-TERM LAKE NETWORK

⊗ 1 BAPTISTE LAKE
⊗ 2 ETHEL LAKE
⊗ 3 NAKAMUN LAKE
⊗ 4 WABAMUN LAKE

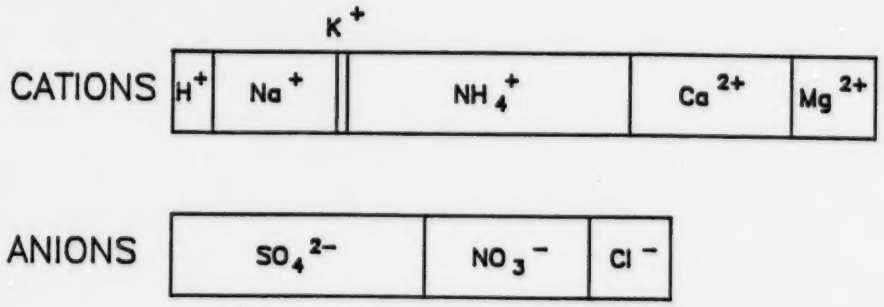
NORTHERN LAKES

△1 LEGEND
△2 UNNAMED
△3 UNNAMED

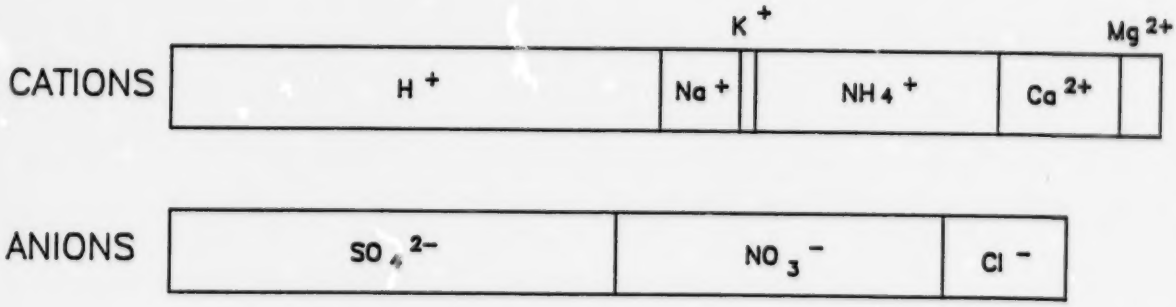
Long Term River and Lake Monitoring Sites

Figure 4

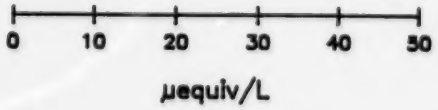
WESTERN PROVINCES



LOWER GREAT LAKES



SCALE



Volume weighted mean values for major ions in wet precipitation (after Munger and Eisenreich, 1983)

Figure 5

relationship cannot be extrapolated to the western provinces because of the presence of neutral sulphate compounds in wet deposition (Lau 1982; Sandhu and Blower 1986, Bertram et al. 1987, Legge 1988a). Therefore, the generation of a critical loading based on wet sulphate deposition is not appropriate for Alberta and perhaps for other prairie provinces.

Effective acidity (EA) is one alternate concept which can be used theoretically to quantify the impact of atmospheric deposition on the acidification of a receptor ecosystem. To apply this concept, three components must be quantified:

1. The actual mineral acidity, expressed as hydrogen ion concentration [H^+], in deposition;
2. The amount of hydrogen ions generated as a result of the receptor-processing of acid-producing substances (A) in deposition;
3. The amount of hydrogen ions lost as a result of the receptor-processing of acid-consuming substances (B) in deposition.

The general equation for these relationships can be written as follows:

$$EA = [H^+] + \sum k_{A_i} \cdot A_i - \sum k_{B_j} \cdot B_j \qquad (3)$$

where k_{A_i} and k_{B_j} are receptor-determined coefficients for i and j processes, respectively. Deposition is expressed as kiloequivalents of hydrogen ion per hectare per year.

The processes affecting the hydrogen ion balance are diverse (Table 6) and vary among ecosystems. For example, the uptake and metabolism of nitrate in soils (denitrification) results in a consumption of hydrogen ions. In contrast, the oxidation of ammonium to nitrite and nitrate (nitrification) produces hydrogen ions. The exact quantitative relationships vary among different ecosystems (aquatic versus terrestrial) and pathways.

Coote et al. (1982) have used this approach for determining soil effective acidity in eastern North America, based primarily on the influence of soil nitrogen cycling on acid production. Assumptions were made about the extent of nitrification, denitrification, and leaching in soils. It was assumed that 50% of the nitrogen deposited to the soil as ammonium was nitrified, and of this quantity 30% was leached and 10% lost by denitrification. The net effect was that the soil would derive 1.15 H^+ ion for each NH_4^+ ion deposited from precipitation. They also estimated that 70% of the nitrate in precipitation would be taken up or denitrified, and the remaining 30% lost through leaching. Thus, 0.7 hydrogen equivalents would be consumed (neutralized) for every nitrate equivalent input to the system. Their equation is of the form:

$$EA = [H^+] + 1.15 \cdot [NH_4^+] - 0.7 \cdot [NO_3^-] \qquad (4)$$

Further, they assumed that the estimated eastern Canada dry deposition rate of 40 percent of wet deposition (for sulphur; Whelpdale and Galloway 1979) was applicable to the effective acidity calculated for all forms of sulphur (and nitrogen) in wet deposition.

Bloom and Grigal (1985) also employed the same formulation for

soil effective acidity, although the coefficients for the NH_4^+ and NO_3^- terms were taken as unity. Turchenek and Abboud (1988) described the use of the entire Bloom and Grigal (1985) soil ecosystem acidification model (Section 4.2 below) but used the wet acidity equation of Coote et al. (1982) combined with a term for dry deposition, i.e.:

$$EA = \text{wetfall } (H^+ + 1.15 \cdot NH_4^+ - 0.7 \cdot NO_3^-) + \text{dryfall sulphur (aerosol + } SO_2) \quad (5)$$

The dry deposition term was taken from sulphur modelling estimates provided by Kociuba (1984) for Alberta, and stoichiometrically converted to hydrogen ion equivalents.

In Ontario, Scheider and Dillon (1982) used a similar approach and defined the total deposition of effective acidity as:

$$EA = [H^+] + [NH_4^+] - k \cdot [NO_3^-] + SO_2 \text{ deposition} \quad (6)$$

where $k = 1$ for aquatic systems

$k < 1$ for terrestrial systems

Sulphur dioxide deposition was estimated from other studies in eastern Canada as a fraction (0.3) of total sulphur deposition.

Note that the gaseous oxides of sulphur and nitrogen (as dry deposition) and alkalinizing components of both wet and dry deposition

should be specifically addressed in future refinements of equation (3). The degree to which receptor processes cause atmospherically deposited sulphur, nitrogen and other substances to contribute to acidification depends on how much of each substance is incorporated into the biological system and how much is removed by other processes such as leaching. This has to be accounted for by using receptor-specific coefficients and ultimately should be quantified for the individual ecosystem in question.

Brydges and Summers (1989) proposed that the value of ($[SO_4^{=}]$ - $[Ca^{++} + Mg^{++}]$) in precipitation is a suitable way to describe the acidifying potential (AP) of wet deposition. They observed that in eastern North America the AP of precipitation varied from 20 to 85% of total sulphate, the remainder of the sulphate being neutralized H_2SO_4, sulphate from salts in dust, or from sea salt. The AP ranged from 20% to 80% of the H^+ in the wet deposition. The rest of the H^+ was contributed by the net effect of nitrogen compounds.

The authors proposed that ammonium and nitrate ions from NH_3 and NO_x emissions would not represent a net acidic loading to the terrestrial environment if they were taken up by vegetation. However, they noted that when nitrogen leaches from the watershed in the form of nitrate, it represents an acidifying demand on the ecosystem. Therefore the overall "net" acidifying potential (NAP) applied to the terrestrial ecosystem is defined by the value of: $[SO_4^{=}]$ - $[Ca^{++} + Mg^{++}]$ in precipitation + $[NO_3^-]$ in runoff from the watershed. This AP is expressed in hydrogen equivalents, and when combined with precipitation rate provides a loading estimate.

4. MODELLING ACIDIFICATION IMPACTS

The necessity of conducting the deposition limit analysis in terms of H^+ equivalents, and relating this H^+ deposition to ecosystem response via the effective acidity or acidifying potential concepts has been outlined in the preceding section. However, the lack of acid-impacted ecosystems in Alberta makes it impossible to generate regional empirical relationships based on H^+ deposition and receptor response, and thereby to derive acceptable deposition limits. The only alternative is to employ models derived elsewhere and modify the input variables in terms of H^+ deposition.

4.1 MODELLING SURFACE WATER ACIDIFICATION

Conceptual, empirical, and complex ecosystem models have been developed to evaluate the acidification status of surface waters (reviewed by Telang 1987). System models are not considered here because the data requirements for running these models limit their general utility to all but specific, well-defined ecosystems. Conceptual models are useful for illustrating ecosystem interactions and general responses to acid loadings. These kinds of models do not require real data and thus have little predictive capability. Empirical models incorporating relationships among key variables are particularly useful for management purposes. However, it is important to fully understand the assumptions and limitations of these relationships when applying them to evaluate acidification impacts.

Empirical models that have been used to assess surface water

sensitivity to acidic deposition and for prediction of acidification impacts are listed in Table 7. The models which have been applied to evaluate Alberta surface waters are also indicated. Although the models are basically derived from the alkalinity/electroneutrality concepts, they are not universally applicable. Modifications are required to account for regional differences in surface water quality conditions, and precipitation chemistry and quantity.

The close relationship between total alkalinity concentration and the sum of calcium plus magnesium concentrations for various mineral lakewaters forms the basis of one such model (Almer et al. 1978). This approximate 1:1 relationship has been documented for lakes that are free from acid effects and low in organic content. Total alkalinity (represented mainly by the bicarbonate ion (HCO_3^-)) is produced by the carbonic acid weathering of the more soluble calcium- and magnesium-based minerals found in rocks and soils. The exported bicarbonate is accompanied usually by an equivalent amount of bivalent Ca^{++} and Mg^{++} cations in runoff and seepage.

During acidification, the HCO_3^- anion is neutralized by the H^+ cation deposited as sulphuric acid on soils and surface waters, with the result that sulphate often becomes the dominant anion in acidic lakewaters. In addition, the Ca^{++} and Mg^{++} cations are exported from watersheds to lakes in larger proportions as a result of ionic exchange against the H^+ and NH_4^+ deposited in acidic precipitation. The result of both processes is a shift away from the 1:1 relationship between total alkalinity and the sum of Ca^{++} plus Mg^{++} in the position of lakes plotted on the Almer et al. (1978) model. This approach was tested

Table 7. Summary of empirical models for evaluating surface water acidification.

	VARIABLES				
	Acidic Deposition	Alkalinity	pH	Watershed Processes	Other
Almer et al. (1978)[1]		Alk, Ca + Mg	-	-	-
Baker and Brezonik (1988)	sulphate, nitrate (as H^+)	Alk	+	+	in-lake alkalinity generation, nitrate, sulphate, hydrology, groundwater, ammonium
Gibson et al. (1983)	sulphate, pH	Ca + Mg + Na + K	+	-	lake sulphate and nitrate
Henriksen (1980)[1]	sulphate, pH	Ca, Ca + Mg	+	-	lake sulphate
Wright et al. (1984)	sulphate (wet + dry)	Ca + Mg	+	neutralization	-
Jones et al. (1984)	sulphate (wet + dry)	Ca + Mg	+	neutralization	-
Nichols and Roberts (1985)	sulphate (natural + emission)	Ca + Mg	+	neutralization	-
Small and Sutton (1986)	sulphate (total)		+	+	organic anions
Dupont and Grimard (1986)[2]	sulphate (volume weighted)	Ca + Mg	+	-	-
Dupont and Grimard (1987)	sulphate (volume weighted)	Ca + Mg	+	-	organic anions, aluminum

Models applied to Alberta lakes

[1]Erickson (1987)
[2]Trew (1988)

by Erickson (1987) to evaluate the acidification status of Alberta lakes.

Henriksen (1980) proposed a similar relationship based on calcium and pH to distinguish between lakes in acidified and unacidified areas. Calcium levels can provide a good approximation for alkalinity in mineral, carbonate-buffered systems and pH is a measure of acidity. Acidified lakes were characterized by high calcium and low pH. This approach is not valid for systems dominated by cations other than calcium or those with non-carbonate buffering (organic acids, metal hydroxides). This limitation was noted by Erickson (1987) for a number of Alberta lakes. More elaborate formulations have been proposed which include watershed geology and corrections for sea salt influences (ESSA 1987; Telang 1987).

The predictive nomogram developed by Henriksen (1982) represents another technique for determining acidification status of lakes. Its major innovation was that it incorporated precipitation acidity (expressed as pH or sulphate) or lake sulphate concentration (a surrogate for acidity) with lake buffering capacity to indicate whether the system is acidic, or acidifying. Erickson and Trew (1987) have illustrated its diagnostic use and its constraints for Alberta lakes.

A number of predictive loading models have been reviewed for use in the Alberta context. The predictive approaches developed to relate acid loadings to changes in surface water quality conditions are similar in that they involve assessment of system responses (alkalinity) to acid inputs.

Dupont and Grimard (1986) proposed a descriptive model for surface water acidification relating precipitation acidity, watershed sensitivity, and resulting lake acidity (pH). Watershed sensitivity was based on lake calcium plus magnesium concentrations (i.e., alkalinity). Sulphate in precipitation was used to represent acid loadings.

Trew (1988) selected and modified the model proposed by Dupont and Grimard (1986) for application to Alberta lakes. Significant modifications included replacement of atmospheric sulphate with an expression of hydrogen ion loadings ($keq(H^+) \cdot ha^{-1} \cdot y^{-1}$) and substitution of alkalinity for calcium plus magnesium (as the watershed sensitivity index). These changes more accurately reflect in-lake conditions and precipitation characteristics in Alberta. Acceptable acid loadings can be derived from this relationship given criteria for the response variable (pH). Note that the model represents observed conditions and not necessarily final steady-state.

More recently, Dupont and Grimard (1987) developed a comprehensive deterministic model for lake acidity based on the work of Small and Sutton (1986). Lake acidity was related to the mean annual volume-weighted sulphate concentration in precipitation, a measure of mineralization corrected for the influence of organic anions, and the partial pressure of carbon dioxide. The model was calibrated against data for 1091 lakes. A 317-lake data set was used for verification purposes. Predictions of in-lake pH levels were in good agreement to measured values. However, additional validation of the model is required. It should be noted that the final forms of the model equations

are not the theoretical solutions but empirical formulations derived for practical purposes.

Inclusion of organic anions reduces the uncertainty associated with predictions of lake acidity in coloured, soft water systems. Muskeg-type lakes generally have a lower pH than mineral-water lakes. Buffering by organic anions is thus implicitly embodied in the model. Buffering by metal hydroxides (i.e., aluminum) at low pH was not considered. This new model could be easily modified and applied to Alberta lakes with the assumptions and alterations employed by Trew (1988) on the initial descriptive model of lake acidification proposed by Dupont and Grimard (1986).

Baker and Brezonik (1988) recently reported the refinement and calibration of a dynamic model for assessing acidification of seepage lakes (Baker et al. 1985). The model was configured to include in-lake processes that both generate (sulphate reduction and nitrate immobilization) and consume alkalinity (immobilization of ammonium) in addition to mineral weathering and acidic deposition. In-lake processes regulating alkalinity were found to be significant in coloured water lakes with long residence times (>5y). Alkalinity generated from the photodecomposition of organic acids and precipitation of humic acids may also play an important role in regulating the hydrogen ion balance of muskeg-type lakes. Although the data requirements for the model are relatively modest, additional calibration of rate coefficients would be required. However, adaptation of the model to predict acidification impacts and to examine groundwater interactions in sensitive Alberta lakes should be considered.

4.2 MODELLING SOIL ACIDIFICATION

The approaches taken to evaluate soil acidification are similar to those used for assessing impacts of acidic loadings on surface waters. Key response variables are identified and related in an empirical fashion to acid inputs. Major soil parameters affected by acid loadings include pH and alkalinity, base saturation, plant available nutrients, and soluble aluminum concentrations. Changes in these constituents also impact on surface water quality conditions.

A number of available empirical and system models were recently reviewed by Turchenek et al. (1987a) and Turchenek and Abboud (1988) in order to select one for application to Alberta soils. The Bloom and Grigal (1985) model was considered the most appropriate. The model is based solely on chemical properties of the soil and soil solution. Biological processes were not explicitly considered. Further, no groundwater interactions are included in the present version of the model.

The model was configured to simulate changes in soil pH, exchangeable bases, and soil solution aluminum concentrations over time for different levels of acidic inputs (Turchenek and Abboud 1988). Only acidification impacts on the top 25 cm of undisturbed native soils were evaluated with the model. Agricultural soils were excluded because agricultural land management practices may have a much greater impact on soil properties than would acidic deposition.

Empirical relationships were used to calculate the soil solution pH, soil aluminum levels, pH, and base saturation. The soil column was taken to be physico-chemically homogeneous and the initial measured

properties reflected a steady-state condition for all relevant processes. All acid loadings were consumed in the soil column. Water lost from the soil column was equal to the incident precipitation minus evapotranspiration.

The following information was required as input: annual rates of precipitation and evapotranspiration, acidity of wet and dry deposition (expressed in equivalents), soil pH, cation exchange capacity, base saturation, and soil partial pressure of carbon dioxide. The quantity and quality of data required for running this model were relatively modest compared to the data requirements for more complex system models.

Turchenek and Abboud (1988) used the model to predict the effects of acidic deposition on changes in the chemical properties of eight different soil types over time. The model was then used to derive deposition rates that cause a specified change in soil pH and base saturation for different simulation periods. Eight Alberta soils with different physical characteristics and chemical properties were evaluated with the model. These soils were selected to cover a representative range of sensitivities to acidification.

The assumptions and limitations of the model were clearly stated and further deficiencies identified by Turchenek et al. (1987a) and Turchenek and Abboud (1988). Many processes were reduced and simplified or simply ignored because they were considered unimportant in the original configuration of the model. Coefficients in the model were not derived specifically for Alberta soils. Further, displacement of basic cations by hydrogen ions is assumed complete and unaffected by other

acid-base reactions or chemical conditions. Additional fine-tuning of the Bloom and Grigal (1985) model for prediction of acidic impacts on Alberta soils merits consideration.

5. LIMITS FOR ACIDIC DEPOSITION

5.1 PROPOSED DEPOSITION LIMITS

The range of deposition limits for lakes, as derived from the Dupont and Grimard (1986) model, is presented in Table 8. Note that the range of alkalinity class intervals for the modified Dupont and Grimard model extended only to 15 mg/L or 300 µeq/L. However, the alkalinity class intervals used for mapping lake sensitivities in Alberta (Table 3) extended to > 40 mg/L. Consequently, deposition limits can be provided only for the highly sensitive lakes (0 to 10 mg/L) at this time.

The alkalinity concentration range for highly sensitive lakes was used to interpolate a critical loading range to a minimum acceptable pH of 6.0. This pH value was proposed as a reasonable threshold value for the protection of most aquatic biota (ESSA 1987), although both the Alberta Surface Water Quality Objectives and the Canadian Water Quality Guidelines have been set at pH 6.5. This approach generates deposition limits ranging from 0.1 to 0.3 keq(H^+)•ha^{-1}•y^{-1}.

For the soils analysis Turchenek and Abboud (1988) estimated acidic loadings that would cause a 10% reduction in base saturation over 100 years for eight soils of low to high sensitivity (Table 9). These loadings range from 0.1 to 1.0 keq(H^+)•ha^{-1}•y^{-1}, with those for highly sensitive soils ranging from 0.1 to 0.3 keq(H^+)•ha^{-1}•y^{-1}.

Table 8. Sensitivity class[1] and proposed acidic deposition limits for surface waters in Alberta.

SENSITIVITY CLASS	ALKALINITY (mg/L)	LOADING $keq(H^+) \cdot ha^{-1} \cdot y^{-1}$
low	>21	> 0.3
medium	11-20	> 0.3
high	0-10	0.1 to 0.3

[1]NRCC (1981)

Table 9. Estimated loadings required to cause a 10% reduction in base saturation of selected Alberta soils over 100 years (Turchenek and Abboud 1988).

SENSITIVITY RATING	SOIL	CLASSIFICATION	LOADING $(keq(H^+) \cdot ha^{-1} \cdot y^{-1})$
low	Cadotte	Solonetzic Gray Luvisol	0.7
	Angus Ridge	Eluviated Black Chernozemic	1.0
medium	Athabasca	Orthic Gray Luvisol	0.4
	Cavendish	Orthic Brown Chernozemic	0.3
	Red Willow	Orthic Black Chernozemic	0.4
high	Mildred A	Eluviated Dystric Brunisol	0.1
	Mildred B	Eluviated Dystric Brunisol	0.2
	Culp	Orthic Gray Luvisol	0.3

It is important to note that within the "highly" sensitive categories progressive refinement of deposition limits would be possible. For instance, the limit for a lake with an alkalinity of 2-3 mg/L would be much less than a lake with 9-10 mg/L, although in this preliminary evaluation both are considered as "highly" sensitive.

The ranges of acid deposition limits for both soils and surface waters are summarized in Table 10.

5.2 ACIDIC DEPOSITION IN OTHER REGIONS

The above rates can be compared to deposition data from various parts of the world where damage has occurred to aquatic systems. According to Likens et al. (1978) and Overrein et al. (1980), damage has been documented at H^+ deposition rates between 0.5 and 1.25 $keq(H^+) \cdot ha^{-1} \cdot y^{-1}$. Scheider et al. (1979a,b) measured average deposition rates of 0.74 $keq(H^+) \cdot ha^{-1} \cdot y^{-1}$ in south-central Ontario. Whelpdale (1979) reported values of 0.4 to 0.8 $keq(H^+) \cdot ha^{-1} \cdot y^{-1}$ for southeastern Canada. Note that these observed data are for H^+ deposition only. Scheider and Dillon (1982) calculated "effective acidity" rates of 0.95 $keq(H^+) \cdot ha^{-1} \cdot y^{-1}$ for lakes in the Muskoka/ Haliburton area of southern Ontario using equation (6). Using the AP approach, Brydges and Summers (1989) estimated deposition at 0.4 to 0.5 $keq(H^+) \cdot ha^{-1} \cdot y^{-1}$ for impacted areas of southern Ontario.

These empirical observations of damaging acidic deposition suggest that the upper limits of the deposition limits proposed for highly sensitive surface waters and soils (0.3 $keq(H^+) \cdot ha^{-1} \cdot y^{-1}$) should not be exceeded.

Table 10. Proposed limits for acidic deposition in Alberta.

	LOADING ($keq(H^+) \cdot ha^{-1} \cdot y^{-1}$)	
SENSITIVITY	SOILS	SURFACE WATERS
low	0.7 to 1.0	> 0.3
medium	0.3 to 0.4	> 0.3
high	0.1 to 0.3	0.1 to 0.3

Independent analyses of critical sulphur and nitrogen loads for various areas of eastern North America and northern Europe substantiate these proposed deposition limits using the effective acidity approach. An international workshop held in Sweden (Nordic Council of Ministers 1988) provides a summary of proposed deposition limits (expressed as H^+ equivalents) for forest soils and surface waters. Selected data are compared with the proposed Alberta limits in Table 11.

The ADRP program, as summarized by Legge (1988a), provides another perspective on critical deposition limits. In a worst-case scenario, the highest sulphur deposition rates determined in the study (at Crossfield East) were assumed to be loaded (as H_2SO_4) to the lakes of highly sensitive regions. This maximum deposition rate of 27.2 $kg \cdot ha^{-1} \cdot y^{-1}$ (which is equivalent to 0.57 $keq(H^+) \cdot ha^{-1} \cdot y^{-1}$) was predicted to cause damage to the sensitive lakes in question.

Finally, the range of critical loadings proposed for Alberta (0.1 to 0.3 $keq \cdot ha^{-1} \cdot y^{-1}$) are essentially the same as thos proposed by the Western and Northern Canada Interim Acid Deposition Target Loadings Task Group to protect highly sensitive lakes (12.5 to 31 $meq \cdot m^{-2} \cdot y^{-1}$; expressed as acidifying potential) (Western Canada LRTAP Technical Committee 1990).

These latter deposition limits were derived using a different empirical relationship, that of Dickson (1983), which relates observed pH in Norwegian lakes with acidic deposition rates. The interpretation of the proposed limits for Western Canada was supported by the application of the Brydges and Summers (1989) watershed cation production approach, i.e., the maximum acceptable AP is given by:

Table 11. Damaging deposition rates ($keq(H^+) \cdot ha^{-1} \cdot y^{-1}$) and proposed deposition limits

Current Estimates of $[H^+]$ Deposition in Damaged Areas	
1. Likens et al. (1978) New Hampshire	0.7 to 1.2
2. Scheider and Dillon (1982) Muskoka	0.95
3. Overrein et al. (1980) Norway	1.0
4. Whelpdale (1979) Eastern Canada	0.4 to 0.8
Proposed Surface Water Deposition Limits from Other Areas	
1. Dickson (1986) Sweden	0.15 to 0.40
2. Henriksen et al. (1986) Norway	0.40
3. Schnoor and Stumm (1987) Switzerland	0.20 to 0.50
4. Schuurkes (1986) Netherlands	0.25
5. Harriman (1988) Scotland	0.20 to 0.40
Proposed Surface Water Deposition Limit for Alberta	
1. Alberta Environment	0.1 to 0.3

$$AP = R \cdot (C - x)$$

where R = annual runoff

C = initial alkalinity or cation concentration

x = minimum acceptable alkalinity (as derived from minimum acceptable pH)

5.3 CURRENT ACIDIC DEPOSITION LEVELS IN ALBERTA

The existing levels of effective acidity and acidifying potential in Alberta have been estimated in order to place the deposition limits proposed in Section 5.1 in a preliminary perspective. Two sources of data have been used: the Alberta Environment provincial data base and the ADRP program results.

The precipitation monitoring network maintained by Alberta Environment was the source of H^+, NH_4^+, and NO_3^- data for the years 1978 to 1987. The ADRP results for the years 1985 to 1987 were also assembled, and wet effective acidity was calculated for each year using the formula of Coote et al. (1982). Average values for each program were subsequently determined (Table 12).

Although not shown, the relative contribution of hydrogen ion to the wet deposition effective acidity is often small. According to these calculations, the potential contribution from wet ammonium is the most important component. Ammonium is greatest at Calgary, Red Deer, Edmonton, and Suffield. Note that deposition of ammonium in European agricultural zones has been shown to cause soil acidification (Van Breemen et al. 1982), and the acidification of lake water through the experimental addition of ammonium (as NH_4Cl) has been demonstrated by Schindler et al. 1985.

Table 12. Recent acidic deposition levels in Alberta measured as "Effective Acidity".

	ACIDIC DEPOSITION (in keq(H^+)•ha^{-1}•y^{-1})		
	Wet Deposition	Dry Deposition	Total Deposition
Alberta Stations[1]			
Beaverlodge	0.11		
Calgary	0.11		
Coronation	0.04		
Edmonton	0.14		
Edson	0.05		
Fort McMurray	0.00		
Lethbridge	0.06		
Red Deer	0.15		
Rocky Mountain House	0.08		
Suffield	0.17		
Whitecourt	0.06		
ADRP Stations[2]			
Fortress Mountain	0.17	0.08	0.25
Crossfield West	0.21	0.30	0.51
Crossfield East	0.20	0.37	0.57

[1] Alberta Environment, unpublished data (1978-1987)
[2] Legge (1988b)

The only estimates for dry deposition are from the ADRP sites. Dry EA was calculated according to Coote et al. 1982 (with H^+ excluded).

The results are summarized in Table 12. Dry deposition is estimated to be the larger component of total acidic deposition at the Crossfield sites, which are on the prairies. This trend is consistent with previous evaluations of dry versus wet sulphur deposition in Alberta (Kociuba, 1984). The apparent differences in wet deposition EA between the Alberta Environment and ADRP data sets may reflect methodological differences in collection techniques (A.E. = monthly bulk; ADRP = event).

The total acidic deposition data estimated in Table 12 span a range of 0.25 to 0.57 $keq(H^+) \cdot ha^{-1} \cdot y^{-1}$, which encompasses the upper limits of the deposition ranges proposed in Table 10.

Acidifying potential was calculated for the 1987 calendar year, again using Ca^{++}, Mg^{++}, and $SO_4^{=}$ data from the Alberta Environment precipitation network. The significance of nitrate leaching in Alberta is not known at this time, and has not been incorporated. The AP values are generally negative, suggesting that acidification is not occurring, and the positive AP values are well below the proposed deposition limits. Note that these estimates are for wet deposition only (Table 13).

These preliminary data point out the need for enhanced monitoring in the highly sensitive regions of Alberta for both wet and dry deposition, and for the refinement of the predictive techniques employed.

Table 13. Acidic deposition in Alberta[1] for 1987 measured as "Acidifying Potential".

	$keq(H^+) \cdot ha^{-1} \cdot y^{-1}$	$meq(H^+) \cdot m^{-2} \cdot y^{-1}$
Beaverlodge	-0.03	-2.66
Calgary	-0.03	-3.17
Cold Lake	+0.004	+0.41
Edmonton	+0.08	+8.06
High Prairie	-0.13	-12.75
High Level	-0.09	-9.31
Fort McMurray	-0.12	-12.33
Kananaskis	-0.31	-31.17
Red Deer	+0.01	+1.21
Suffield	-0.45	-44.57
Whitecourt	-0.07	-6.55

[1] Alberta Environment, unpublished data (1987)

6. SUMMARY

It is recommended that the general relationships (Equation 3) for estimating effective acidity and acidifying potential be further developed for use in Alberta. The relative importance of individual terms and processes must be evaluated and appropriate coefficients derived for Alberta soils and surface waters.

Good quality data for wet and dry deposition in Alberta are required to comprehensively evaluate the proposed deposition limits. A monitoring program for dry deposition employing the best available technology should be established. The present wet deposition monitoring network should be reviewed and greater emphasis given to monitoring sensitive areas.

Monitoring, mapping, and classification of surface waters andsoils should be continued. The sensitivity classification and mapping of all provincial surface waters are not yet complete.

The most pressing requirements are for the further development, refinement and adaptation of empirical models for the prediction of acidification impacts in Alberta. The ranges of the deposition limits proposed in this report remain somewhat large for implementation. The proposed deposition limits should be reviewed and revised as additional information from research activities and monitoring becomes available. Specific information needs are summarized in Table 14.

Table 14. Summary of information needs.

INFORMATION NEEDS
acidic deposition monitoring - reassessment, coordination and monitoring in sensitive areas - implementation of a dry deposition monitoring program employing best available technology
continued monitoring and improved sensitivity mapping of surface waters and soils
development of better predictive capabilities for acidification impacts
spatial analysis and development of regional depositional limits
further refinement of the effective acidity and acidifying potential concepts and assessment of the relative contributions of other acidic/basic substances in atmospheric deposition and watershed runoff
assessment of acidic deposition effects on vegetation
assessment of sediment, shoreline and watershed interactions (alkalinity production) in the acidification of aquatic systems
additional information on peatlands, bogs, and fens is required - distribution, extent, type, chemistry, nutrient cycling, organic cycling, water balance, acid sensitivity
evaluation of the role of vegetation and organic matter in the acidification process of soils and surface waters
assessment and inventory of the acidification status and chemical properties of agricultural soils

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APPENDIX

GLOSSARY OF TERMS

Acid	Any substance that is capable of donating a proton (hydrogen ion)
Acid Neutralizing Capacity (ANC)	A measure of the net deficiency of protons over a reference level of protons, determined from titration with strong acid to a preselected equivalence point
Acidification	A build-up of hydrogen ions above normal levels
Acidity	Proton or hydrogen ion concentration usually expressed in terms of pH (negative logarithm to the base ten of the hydrogen ion concentration; $pH = -\log [H^+]$
Acid Deposition	Transfer of acid and/or acid-forming emissions from the atmosphere to the earth's surface (includes wet plus dry forms)
Aerosol	A suspension of fine solid on liquid particles in gas
Alkalinity	A measure of the acid neutralizing capacity of an aqueous solution (carbonate alkalinity is a measure of alkalinity from carbonate plus bicarbonate)
Anion	A negatively charged ion
Anthropogenic	Man-made or man-modified materials
Base	Any substance that is capable of accepting or gaining a proton
Base Neutralizing Capacity (BNC)	A measure of the net excess of proton over a reference level, equivalent to the sum of all acids that can be titrated with a strong based to an equivalence point
Base Saturation	Proportion of the cation exchange capacity of a soil occupied by base cations (Na^+, K^+, Ca^{2+}, Mg^{2+})

Best Practical Technology	Pollution control or process technology which is technically proven by virtue of current usage or extrapolation and which is affordable within the economic viability of the project.
Buffer	Any substance in solution that will resist changes in pH by donating or accepting protons (usually acid/base pairs such as a weak acid and its conjugate base)
Buffer Intensity	A measure of the tendency of the solution to change pH upon addition of acid as base
Bulk Sample	Sample containing wet plus dry deposition (wet samplers only collect samples during precipitation events whereas bulk samples are left open)
Cation	A positively charged ion
Cation Exchange Capacity (CEC)	A measure of the total buffering capacity of a soil (primarily by clay and organic matter)
Critical Loading	The maximum level of acidic atmospheric deposition that will not cause chemical changes leading to long-term harmful effects on the most sensitive ecological systems
Dry Acidic Deposition	The transport of acids and acid-forming substances to the earth's surface by all processes that do not involve some form of precipitation
Ecosystem	A complex of ecological community and environment forming a functioning whole in nature
Equilibrium	A static of dynamic balance between opposing system components
Infiltration	Downward entry of water into soil
Leaching	Downward movement within the soil of nutrients in solution
Long Term	A period not less than 100 y
Mineral Acidity	A measure of the base neutralizing capacity of an aqueous solution
Minerotrophic	A supply of water to vegetation originally derived from mineral soils or rocks

Neutralize	Consumptive reaction of protons with basic substances
pH	Negative logarithm to the base ten of the hydrogen ion concentration ($pH = \log [H^+]$)
Sensitivity	The degree to which acidic inputs will cause changes in the system
Short Term	A period less than 1 year
Soil Fertility	Soil status in relation to available plant nutrients
Soil pH	Hydrogen ion concentration ($-\log [H^+]$) of a soil at a specified moisture content
Soil Solution	Aqueous liquid phase of the soil and its solutes consisting of ions dissociated from the surfaces of the soil particles and of other soluble nutrients
Steady-State	Condition of constant change without fluctuations, a system at steady is not necessarily at equilibrium but an equilibrium condition is steady-state
Target Loading	The maximum level of acidic atmospheric deposition, that affords long-term protection from adverse ecological consequences and that is practically and politically achievable (distinct from critical loading)
Wet Acidic Deposition	The transport of acids and acid-forming substances to the earth's surface by some form of precipitation (rain, snow, fog)